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Title

Isotopic investigations for the origin of relic from the Matsusaki site, Japan

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Abstract

We measured the environmental radioactivity and isotope ratio, $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, $^{234}\text{U}/^{238}\text{U}$, $\delta^{13}\text{C}$ and, $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio, of adhesion that adhered to a wooden tubing vessel obtained from the Matsusaki site, one of the salt-production sites where Moshio is manufactured, in Aichi Prefecture, Japan. The environmental radioactivity and isotope ratio verified that the adhesion originated from the seaweed and/or sea grass.

Main text

1. Introduction

The intake of salt is necessary and indispensable to human life. The production of salt from other than seawater is dependent on resources such as rock salt and salt lakes. About 180 million tons of salt are produced annually throughout the world, with the salt from seawater making up about 1/4 of the total. In Japan, which has no rock salt and salt lakes, the source of almost all salt is seawater.

The salt that is called “Moshio” was first made in ancient Japan. It has been thought that Moshio had been made from seaweed and seawater. Many relics used for making salt that are called salt manufacturing earthenware have been excavated at salt-production sites. Because the earthenware was thin and easily cracked, it was frequently thrown away when it was no longer useful. In some salt-production sites, a white substance remained adhered to the abandoned the salt manufacturing earthenware. One of the famous salt-production sites where Moshio was manufactured is Matsusaki site, Aichi Prefecture, Japan. A large quantity of salt manufacturing earthenware was discovered at this site. In addition, a wooden tubing vessel was found that was thought to have been used to collect brine. A white substance often discovered in other salt-production sites was found stuck to both the salt manufacturing earthenware and the wooden tubing vessel at the Matsusaki site. Mori [1] analyzed the number of diatom valves of a sample of deposition that adhered to the wooden tubing vessel at the Matsusaki site. The most prevalent diatom was *Cocconeis scutellum*, a marine epiphytic diatom. The detection of marine epiphytic diatoms suggested the manufacture of salt at the Matsusaki site. Furthermore, as the same marine epiphytic diatoms in the earthenware had been discovered in the wooden tubing vessel, it was deemed that the wooden tub had contained liquid that originated from fractionated marine particles.

However, there is no chemical evidence of salt manufacture at the Matsusaki site other than the results of the marine epiphytic diatom analysis.

In our study, environmental radioactivity in the substance that adhered to the wooden tubing vessel was measured to investigate the origin of the substance. Then, there was a search for evidence based on the seaweed or sea grass at the Matsusaki site. The judgment as to whether the substance that adhered to the wooden tubing vessel was of marine origin is discussed from a comparison of environmental radioactivity and chemical analysis. An attempt to determine the origin of the substance was made by examining the environmental radioactivity in the substance:

1) $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, 2) activity ratios of $^{235}\text{U}/^{238}\text{U}$ and $^{234}\text{U}/^{238}\text{U}$, 3) $\delta^{13}\text{C}$, and 4) $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio.

2. Materials

2-1 Adhesion substance obtained from the Matsusaki site

The excavation situation of the substance that adhered on wooden tubing vessel thought to have been brine in the Matsusaki ruins was reported by Tatematsu [2]. The wooden tubing vessel divided the log into half, and it was buried in the sand along the coast. The white deposit that adhered to the wooden tubing vessel was a thick calcareous deposit. A part of this substance in the wooden tubing vessel was used for the analysis.

2-2 Shell

A shellfish fossil is a shell excavated from a layer of pure shell at the Matsusaki ruins. The pure shell layer is composed only of shell, and does not contain ordinary soil. As the shellfish obtained from the pure shell layer is little polluted by soil, the shell was

chosen from pure layer shellfish. The shell used for the analysis was that of a bivalve.

The shell was washed with a brush and ultra-pure water.

3. Methods

3-1 Chemical form and major element of adhesion substance

The adhesion substance was dissolved with 1 M HNO₃. The solution containing the dissolved adhesion sample from the Matsusaki site was filtered with a membrane filter having a pore size of 0.45 μm, and the water was diluted with ultra-pure water. The concentrations of B, Al, P, Ca, Mn, Fe, Sr, and Ba in the sample were determined by ICP-AES, ICPS7500 (Shimadzu Co., Ltd). The chemical composition of the sample was measured by XRD (Rinto 2100, Rigaku Co., Ltd.).

3-2 Separation and measurement of ⁸⁷Sr/⁸⁶Sr ratio

A 0.5 g of the adhesion substance sample was soaked in ultra-pure water for 3 days after which the water was removed from the sample that remained. The remaining sample was soaked in a solution of 1 M CH₃COONa for 3 days, and then any remaining water was extracted. The remaining sample was soaked in a solution of 1 M CH₃COOH containing CH₃COONa adjusted to pH 5 for 7 days, and then carbonate was dissolved into the sample solution. Each sample solution, ultra-pure water, CH₃COONa, and CH₃COOH/CH₃COONa, was filtered with a 0.45 μm membrane filter. Cation exchange resin (Muromachi Chem. Co., Ltd.) was used to extract Sr from the sample water. A shellfish fossil sample was cut with a dental drill, avoiding the surface portion, and only the inside was further cut with the dental drill. To obtain the extracted substance, about 1 mg powder of the shellfish fossil sample was dissolved with 0.1 M CH₃COOH/CH₃COONa buffer solution. The solutions extracted from the

substance were passed through the cation exchange resin, and the Sr in solution was separated. Isotopic measurements were performed using a Thermo Finnigan™ TRITON multi-collector thermal ionization mass spectrometer (TIMS). Data acquisition was at high intensity (10 V) and in static collection mode with 100 scans (10 scans \times 10 blocks). The “virtual amplifier” function of the TRITON TIMS was applied to eliminate all gain calibration errors. Mass fractionation was collected by using $^{86}\text{Sr}/^{88}\text{Sr} = 0.1194$. Throughout the test runs, the average internal precision was ± 0.000009 (2 SE). Replicate measurements ($n = 6$) of NIST SRM 987 yielded an average of 0.710262 ± 0.000010 (2 SD [= external precision]). Normalization of the $^{87}\text{Sr}/^{86}\text{Sr}$ data was based on the difference between the whole-run average of NIST SRM 987 and its recommended value, for which we used 0.710250; i.e., all raw sample $^{87}\text{Sr}/^{86}\text{Sr}$ ratios were normalized by subtracting 0.000012.

3-3 Measurement of $^{234}\text{U}/^{238}\text{U}$ and $^{235}\text{U}/^{238}\text{U}$ activity ratio

A sample of the adhesion substance was dissolved in 2 M HNO_3 and the solution was filtered with filter paper (5A). The solution containing uranium and thorium was loaded onto a U-TEVA resin column (Sowa Trading Co., Inc.), the thorium in the resin was removed, and the remaining resin was rinsed with 2 M HNO_3 . The ^{238}U in the solution was measured with ICP-MS (Yokogawa Co., Ltd.). The uranium isotopes (^{234}U , ^{235}U , and ^{238}U) in the sample solutions were measured by TIMS and $^{234}\text{U}/^{238}\text{U}$ and $^{235}\text{U}/^{238}\text{U}$ ratios were obtained.

3-4 Measurement of $\delta^{13}\text{C}$

The adhesion substance and shell obtained at the Matsusaki site was stored in a glass bottles. The atmosphere that existed on the surface of the adhesion substance and the

shell and in the bottles was removed with He gas. After the atmosphere was evacuated, the adhesion substance and shell were dissolved in hydrochloric acid, generating CO₂ gas. The CO₂ generated from the samples was collected in a glass syringe that separated the CO₂. The CO₂ was introduced into GC-C-IRMS, the ¹²C and ¹³C were measured, giving the δ¹³C value. The δ¹³C is calculated by Pee Dee Belemnite (PDB).

3-5 Measurements of Ra isotopes

Radium isotopes (²²⁸Ra and ²²⁶Ra) in the adhesion sample were packed into an airtight tin canister. The canister was stored for 1 month for γ-ray spectrometry to allow ²¹⁴Pb and ²²⁸Ac to reach radioactive equilibrium with their parents, ²²⁶Ra and ²²⁸Ra, respectively. The radioactivity of ²¹⁴Pb and ²²⁸Ac was determined by 351 keV and 911 keV γ-rays, respectively, using a Ge detector with 40% relative efficiency.

4. Results and discussion

4.1 Chemical composition

Fig. 1 shows the X-ray diffraction pattern of the adhesion substance. The chemical composition of the adhesion sample was primarily composed of calcite (CaCO₃). In addition, it is difficult to imagine that the adhesion substance is evaporated seawater because there is no peak from halite. Concentrations of some of the elements along with a sample of sea algae obtained from the reference value by Yamamoto [3] are shown in Fig. 2. The pattern of the concentrations in the Matsusaki samples is similar to that of the seaweed samples.

4.2. ⁸⁷Sr/⁸⁶Sr ratio in the adhesion substance

The ratios of the $^{87}\text{Sr}/^{86}\text{Sr}$ isotopes in the adhesion substance and the shell obtained from the Matsusaki ruins are shown in Fig. 3. The ratio of $^{87}\text{Sr}/^{86}\text{Sr}$ in the shell reflects that in seawater at the time the shell was collected from the sea. The ratio of $^{87}\text{Sr}/^{86}\text{Sr}$ in the shell reflects that in seawater at the time the shellfish was gathered at the Matsusaki ruins and coetaneous area. The ratio of $^{87}\text{Sr}/^{86}\text{Sr}$ was 0.70920 - 0.70928 for each solution extracted from the adhesion substance to ultra-pure water, the CH_3COONa solvent, and the $\text{CH}_3\text{COOH}/\text{CH}_3\text{COONa}$ solvent, 0.70935 ± 0.00001 for the residue of the adhesion substance, and 0.70918 ± 0.00001 for the shell. Today, the ratio of $^{87}\text{Sr}/^{86}\text{Sr}$ in seawater is 0.709175 ± 0.000006 (Ando, unpublished data). The ratio of $^{87}\text{Sr}/^{86}\text{Sr}$ in the adhesion substance at the Matsusaki ruins consisted of that in the seawater as estimated from the shell. Thus, it was shown that the origin of the adhesion substance was a product of the sea (either seawater or something marine product).

4.3. $^{234}\text{U}/^{238}\text{U}$ and $^{235}\text{U}/^{238}\text{U}$ activity ratios

Table 1 shows the concentration of ^{238}U and the activity ratios of $^{234}\text{U}/^{238}\text{U}$ along with $^{235}\text{U}/^{238}\text{U}$ in the adhesion substance. The concentration of ^{238}U in the adhesion substance is 7.2-7.5 mg/kg. The activity ratios of $^{235}\text{U}/^{238}\text{U}$ and $^{234}\text{U}/^{238}\text{U}$ in the adhesion substance are $0.04601(1\sigma=0.00006)$ and $1.19(1\sigma=0.07)$, respectively. The activity ratio of $^{235}\text{U}/^{238}\text{U}$ in the natural environment except for the area around the Oklo natural nuclear reactors is 0.046 [4]. The $^{235}\text{U}/^{238}\text{U}$ activity ratio in the adhesion sample was within the range of the natural environmental sample. Miyake et al. [5] reported that the $^{234}\text{U}/^{238}\text{U}$ activity ratio in plankton and algae ranged from 1.07 to 1.18, which coincided well with those in environmental seawater. The activity ratio of the $^{234}\text{U}/^{238}\text{U}$ ratio in seawater is 1.14-1.15 [4], [6], which agrees well with the value of the seawater within the range of error compared with the uranium isotopes in the adhesion

substance. The activity ratio of $^{234}\text{U}/^{238}\text{U}$ in the adhesion substance supports the belief that the origin of the adhesion substance is a marine product.

4.4. $\delta^{13}\text{C}$

The possibility that the adhesion substance is a product of sea algae or seaweed was examined by the following techniques. The carbon isotopes ($\delta^{13}\text{C}$) contained in the shell and in the adhesion substance from the Matsusaki ruins were measured, and $\delta^{13}\text{C}$ in the adhesion substance was compared with the seawater at that time, the sea grass, and the seaweed. The $\delta^{13}\text{C}$ values of the shell and the adhesion substance are shown in Fig. 4(a). Because the ratio of the carbon isotopes in seawater is reflected for $\delta^{13}\text{C}$ in the shellfish fossil (shell), $\delta^{13}\text{C}$ in the shell sample shows $\delta^{13}\text{C}$ in seawater at the time of discard into the shell mound, the Matsusaki ruins. Fig. 4(b) is the value of $\delta^{13}\text{C}$ in seaweed, sea algae, river deposits, coastal deposits, and surrounding soil. A value of $\delta^{13}\text{C}$ in the adhesion substance ranged from -10‰ to -7‰ . This was in the range of the sea algae or seaweed, and a value that was smaller than $\delta^{13}\text{C}$ in seawater was indicated. It means that light carbon isotopes is richer than that in seawater. Because the $\delta^{13}\text{C}$ in the adhesion substance was larger than $\delta^{13}\text{C}$ of the river deposit, the coastal deposit, and the surrounding soil, as shown in Fig. 4(a) and (b), it was thought that the $\delta^{13}\text{C}$ in the adhesion substance reflected the value of a marine product. Based on the ratio of the $^{87}\text{Sr}/^{86}\text{Sr}$ in the adhesion substance, the material of the adhesion substance did not originate from the land but from seawater or seaweed. Furthermore, as the value of $\delta^{13}\text{C}$ in the adhesion substance was smaller than that in the seawater, the source of the adhesion substance would be seaweed or sea grass. This interpretation followed from the discovery of the marine epiphytic diatoms from the wooden tubing vessel at the coast [1].

4.5. $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio

Next, contradiction of the interpretation about the origin of the sample based on the observed ^{13}C in the sample was confirmed by measuring the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio. Because the ratio of the Sr isotope and the $^{234}\text{U}/^{238}\text{U}$ ratio in the adhesion substance showed that the substance was a marine product, the origin of the adhesion substance has to be either seawater or something marine product. Here, assuming that the origin of the adhesion substance is only seawater, when the seawater had been isolated from sea, as the amount of ^{226}Ra and ^{228}Ra that is contained in the isolated seawater decreases over time, the ^{226}Ra and ^{228}Ra in the adhesion substance similarly decrease over time. The Matsusaki site is facing on the Pacific Ocean. The concentrations of ^{228}Ra and ^{226}Ra in the surface seawater located along the coast of the Pacific Ocean of Japan are 0.2-1.4 mBq/l and 1.1-1.9 mBq/l, respectively, and the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio is 0.2-0.8 [12], [13], [14]. Fig. 5 (a) shows that the relationship of the concentrations of the ^{228}Ra and ^{226}Ra and $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratios of the seawater samples were isolated from the sea. The initial values of the concentrations of ^{226}Ra and ^{228}Ra shown in Fig. 5 (a) are 0.4 mBq/l and 1.2 mBq/l, respectively, representing the concentrations of ^{226}Ra and ^{228}Ra contained in the seawater at the time humans were active at the Matsusaki site about 1000 years ago. As the half-life of ^{228}Ra is 5.8 years, the amount of ^{228}Ra in seawater isolated from the sea will fall below the detection limit. Therefore, it is thought that the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio of the seawater isolated from the sea would fall below the detection limit after 1000 years or more. Fig. 5 (b) shows the $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in the adhesion substance along with that in the seawater, river water, and sedimentary rock [13], [16], [17]. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in the adhesion substance is 0.59 ± 0.11 , as shown in Fig. 5 (b). It is on the same order of the seawater

that is in the Pacific Ocean around the islands of modern Japan. This supports the idea that the origin of the adhesion substance is not from seawater alone. Assuming that the adhesion substance was made only from seawater, as seawater contains NaCl, the data of the chemical form that was primarily composed of calcite and not halite is contradicted. The content of U and Th in seawater are, respectively, $2.6 - 3.7 \times 10^{-6}$ g/l [5] and $2 - 91 \times 10^{-10}$ g/l [6], [17], [18] on the orders of magnitude of an ultra low-level. Seaweed and/or sea grass is able to concentrate much U and Th from seawater. It is thought that ^{228}Ra in the adhesion substance was produced from ^{232}Th . Therefore, the origin of the ^{232}Th in the adhesion substance might be seaweed and/or sea grass. Because the adhesion substance contains ^{232}Th , its origin is supposed to be marine product took part. The explanation that the adhesion substance originated from seaweed and/or sea grass is not contradicted by the value of $\delta^{13}\text{C}$ in the substance. The activity ratio and environmental radioactivity support the idea that the adhesion substance originated from seaweed and/or sea grass.

5. Conclusion

The $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in the origin of the adhesion substance obtained at the Matsusaki site was 0.70920 -0.70928. This value agreed well with the estimated value of seawater from shell, showing that the adhesion could originate from seawater and/or marine products. The activity ratio of $^{234}\text{U}/^{238}\text{U}$ showed the values of the seawater and/or marine products as well as the $^{87}\text{Sr}/^{86}\text{Sr}$ ratio, and this explanation based on environmental radioactivity supported that the origin of the adhesion substance was seawater and/or a marine product. Furthermore, the $\delta^{13}\text{C}$ in the adhesion substance was in a range that was smaller than the value of seawater and larger than that of a land plant. It was suggested that the adhesion substance could originate from not only seawater but

also seaweed and/or sea grass. The $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in the substance showed that the adhesion substance originated not only from seawater but also from seaweed and/or sea grass.

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Figures captions

Figure 1: X-ray diffraction pattern for the adhesion.

C: Calcite

Figure 2: Concentrations of elements in the adhesion along with seaweed sample.

○: adhesion, ●: Seaweed

Figure 3: $^{87}\text{Sr}/^{86}\text{Sr}$ ratio in the each solutions extracted from the adhesion and shell obtained from Matsusaki site.

○: Adhesion, △: Shell, ●: Seawater

Figure 4: (a) $\delta^{13}\text{C}$ in the adhesion and shell obtained from Matsusaki site.

(b) $\delta^{13}\text{C}$ in environmental samples.

○ : Adhesion, ●: Shell (seawater),

(1) Kuramoto and Minagawa (2001), (2) Mito et al. (2007),

(3) Yamamuro et al. (2004), (4) Ishihi et al. (2001), (5) Shimoda et al. (2007).

Figure 5: (a) Relation between $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio of seawater isolated from sea and time. (b) $^{228}\text{Ra}/^{226}\text{Ra}$ activity ratio in the adhesion along with coastal water, river water and sedimentary rock.

1) Nakano-Ohta and Sato (2006), (2) Yamada and Nozaki (1986),

(3) Ohta et al. (2008), (4) Nakano-Ohta and Sato (2005), (5) Nakano-Ohta et al. (2005).

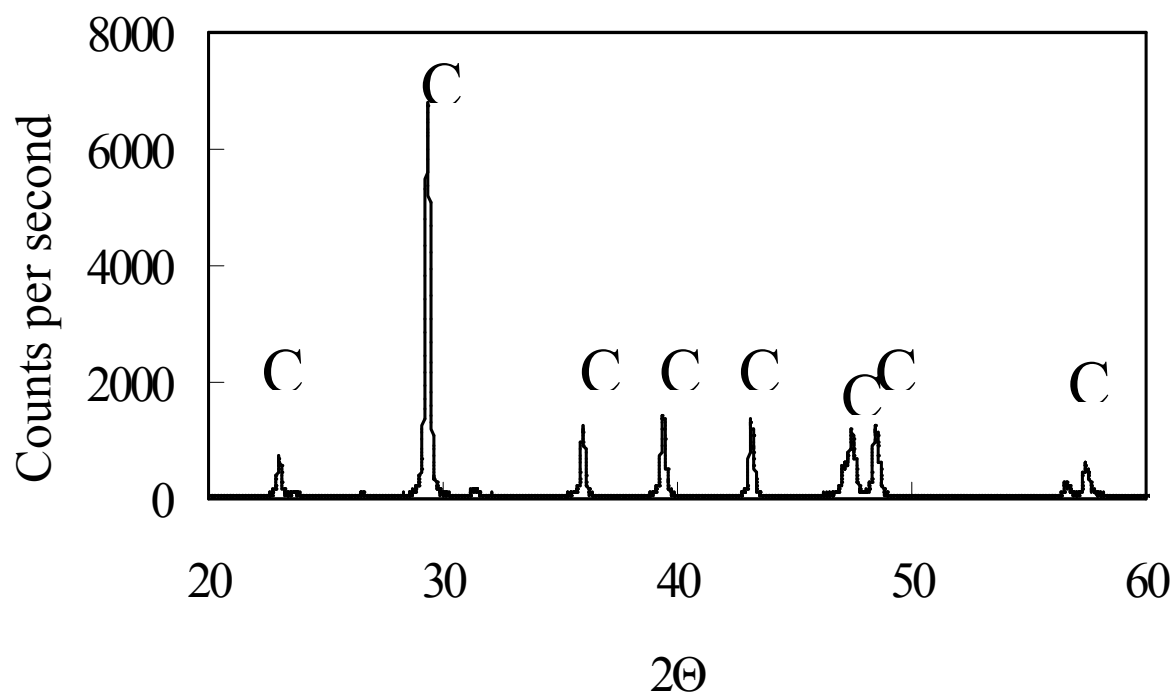


Fig. 1

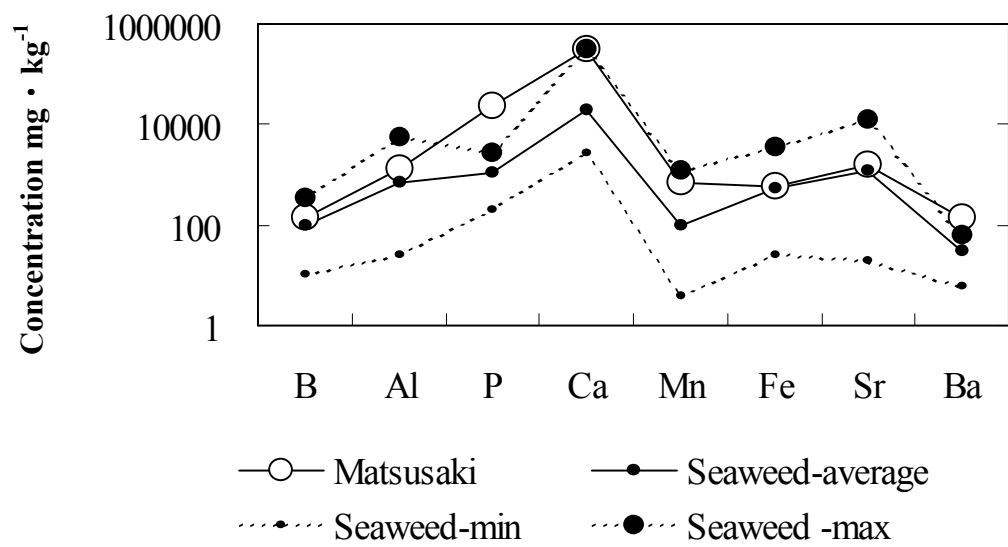


Fig. 2

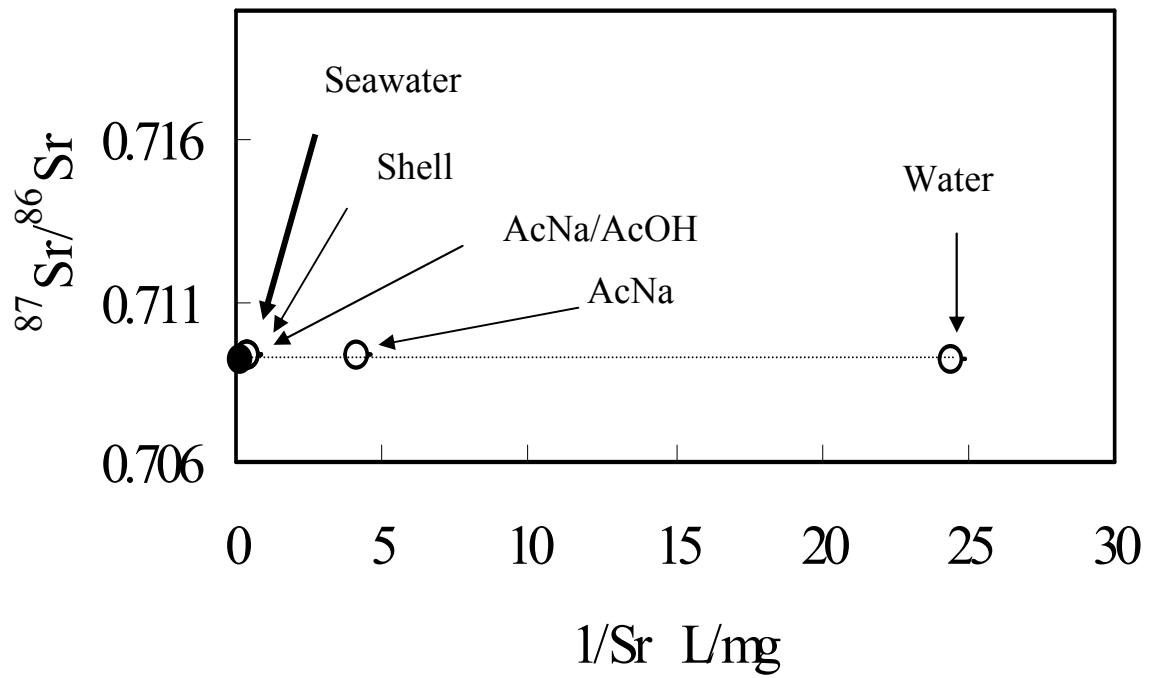


Fig. 3

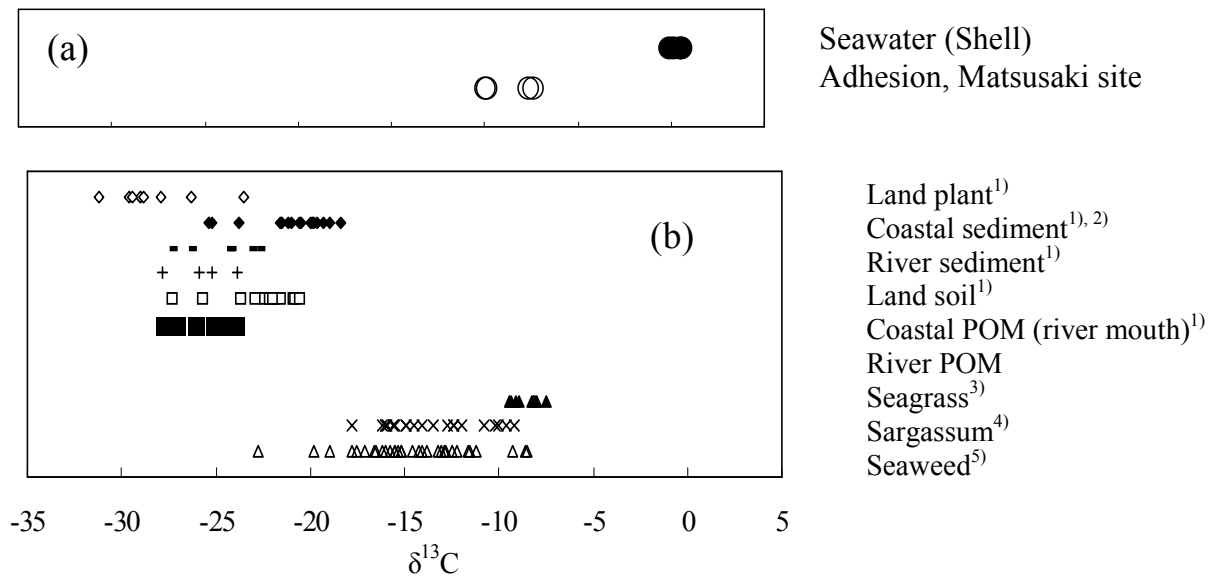


Fig. 4

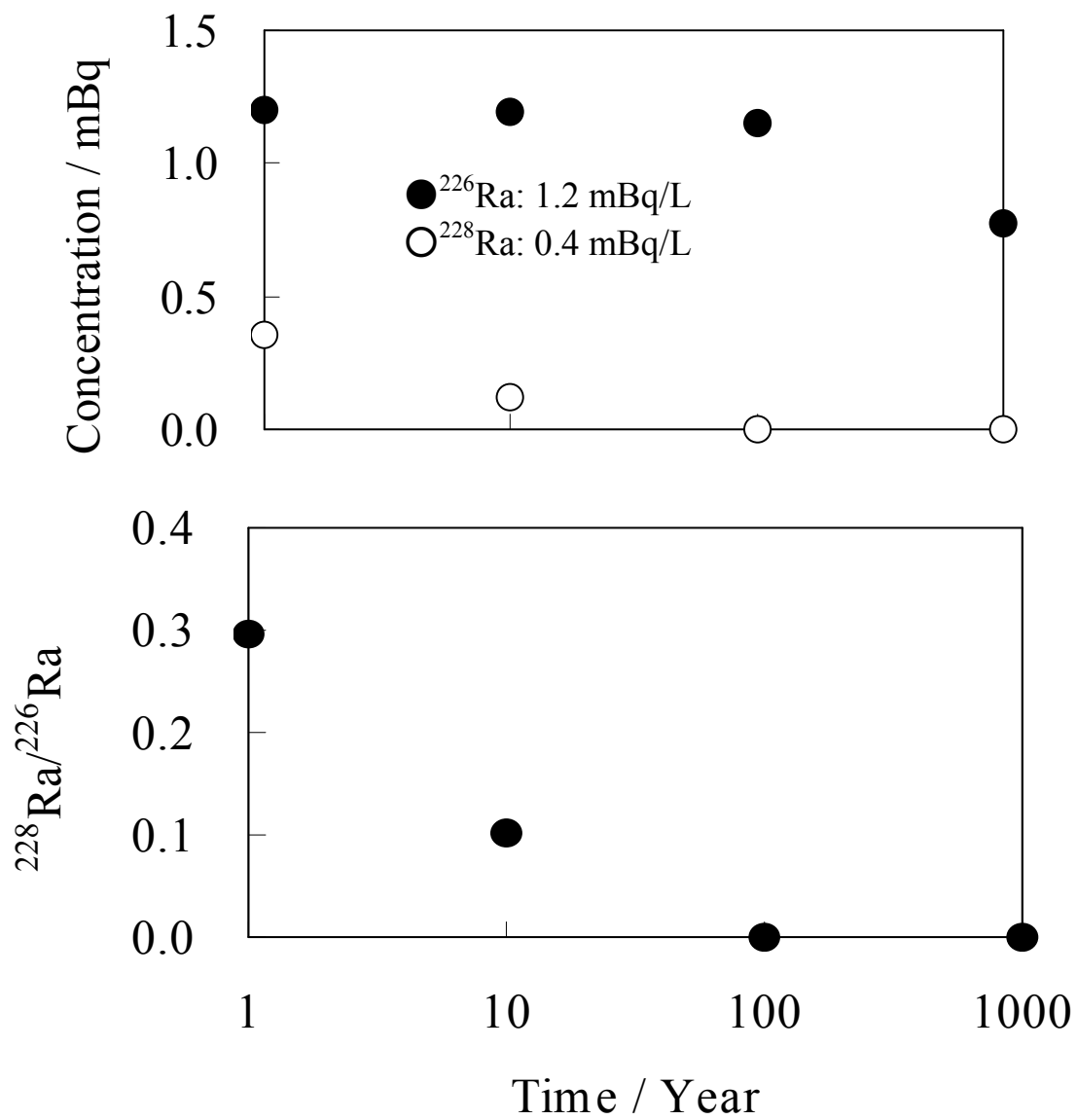


Fig. 5(a)

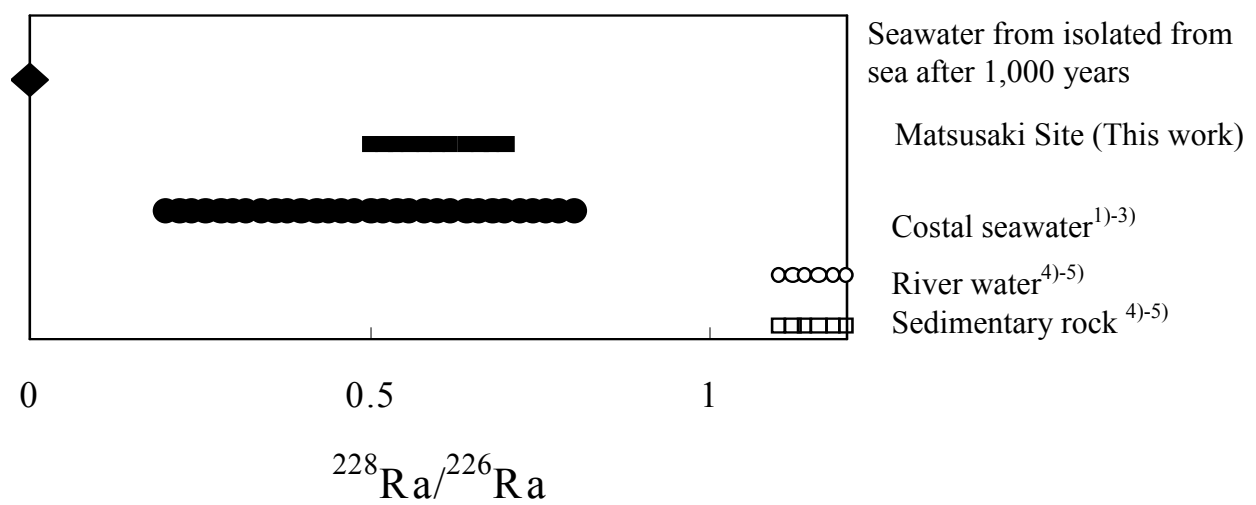


Fig. 5(b)

Table 1 Concentration of ^{238}U , and ratios of $^{235}\text{U}/^{238}\text{U}$ and $^{234}\text{U}/^{238}\text{U}$ in Matsusaki sample along with seawater

^{238}U concentration	$(^{235}\text{U}/^{238}\text{U})$ activity ratio	$(^{234}\text{U}/^{238}\text{U})$ activity ratio
7.3 – 7.5 mg/kg	0.04601 ($2\sigma=0.00006$)	1.19 ($2\sigma=0.07$)
Seawater	0.045-0.046**	1.14-1.15*· **

*[5], **[4].